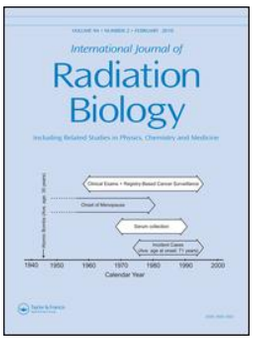

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ORIGINAL ARTICLE

MPS dose reconstruction for internal emitters: some site-specific issues and approaches

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ABSTRACT

Background: As part of the Million Person Study (MPS), dose reconstructions for internal emitters have been performed for several U.S. facilities where large quantities of radionuclides were handled. The main challenges and dominant sources of potential error in retrospective dose estimates for internally exposed workers have been found to vary from site to site. This article discusses some important issues encountered in dose reconstructions performed for selected MPS sites and the approaches used to address those issues. The focus is on some foundational components of retrospective dose assessments that have received little attention in the literature.

Methods: The discussion is built around illustrative exposure data and dose reconstructions for workers at selected facilities addressed in the MPS. Related findings at some non-MPS sites are also discussed.

Results: Each of the following items has been found to be a major source of potential error in reconstructed tissue doses for some MPS sites: identification of all dosimetrically important internal emitters; the time pattern of intake; the mode(s) of intake; reliability of bioassay measurements; application of surrogate (coworker) information in lieu of, or in conjunction with, worker-specific monitoring data; the chemical and physical forms of inhaled radionuclides; and the relation of air monitoring data to actual intake.

Conclusions: (1) Much of the dose reconstruction effort for internal emitters should be devoted to development of best feasible exposure scenarios. (2) Coworker data should be used to assign exposure scenarios or dose estimates to workers with missing exposure data only if there is compelling evidence of similar coworker exposure. (3) Bioassay data for some radionuclides and periods of operation at MPS sites are of questionable reliability due to sizable uncertainties associated with contamination, recovery, or background issues. (4) Dose estimates derived solely from air monitoring data should be treated as highly uncertain values in the absence of site-specific information demonstrating that the data are reasonably predictive of intake. (5) For intakes known or assumed to be via inhalation, the uncertainty in lung dose typically is much greater than the uncertainty in dose to systemic tissues, when dose estimates are based on urinary excretion data. (6) The lung dose estimate often can be improved through development of site-specific respiratory absorption parameter values. (7) There is generally insufficient site-specific information to justify development of site-specific systemic models.

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Introduction

As part of the Million Person Study (MPS), dose reconstructions have been performed for several U.S. facilities where large quantities of radionuclides were handled. Operations at some of these facilities began in the 1940s and continued for several decades, and some of the facilities are still in operation. Thousands of workers at these facilities were exposed to elevated levels of external radiation, and a generally smaller but considerable number of workers had elevated intake of radionuclides, particularly in the early years of operation. The MPS dose reconstructions have provided yearly dose estimates for workers exposed from external or internal sources, as well as broad assessments of the sources and extent of uncertainties in the reconstructed doses.

MPS estimates of external dose to a worker typically are based on personal dosimeter readings recorded over the worker's career. A personal dosimeter reading generally represents an operational dose quantity needed to demonstrate compliance with regulatory limits and involves nontrivial uncertainties when extrapolated to tissue doses (NCRP 2018). Nevertheless, dosimeter readings are expected to provide meaningful and reasonably complete records of external exposure to most radiation workers.

Monitoring data from which to reconstruct doses from internal emitters at MPS sites generally are more uneven and difficult to interpret than data for external radiation sources. Bioassay data are the preferred monitoring data for dose reconstructions, but often are sparse or absent for workers with high potential for intake of radionuclides. Also, interpretation of bioassay data often relies on incomplete information on the nature of the exposure such as the time pattern of intake, mode(s) of intake, and the chemical and

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physical form of the internally deposited radionuclide(s). Air monitoring data are often available for specific work areas, but are crude and often unreliable predictors of the level of intake of airborne radionuclides. In some cases, no monitoring data are available for workers with the potential for elevated intake of radionuclides.

For such reasons, a reasonably complete and meaningful set of retrospective dose estimates for internal emitters for an MPS site requires a carefully designed study built on a wider set of site-specific information than bioassay or air monitoring records. The study should begin with a detailed review of the site history including specific operations, materials handled, incident reports, worker-specific records, and, if feasible, interviews of former workers. This type of information is needed to construct reasonably realistic exposure scenarios for individual workers or groups of workers, to assess the need for site-specific or worker-specific biokinetic models, and to gauge the validity of applying coworker data to potentially exposed workers with sketchy or missing records. A critical review of bioassay measurement techniques over time is also needed to assess the reliability of these data. These early steps in the dose reconstruction process provide the foundation for interpreting monitoring data, selection of appropriate biokinetic models for exposures to individuals or groups, and the feasibility of characterizing site-specific or worker-specific respiratory retention and absorption for forms of radionuclides whose bioassay data are not adequately reproduced by standard models.

This article discusses and illustrates the importance of these foundational steps in the dose reconstruction process in the MPS, some differences between sites in the nature of the information available to develop exposure scenarios or derive dose estimates, and some site- or worker-specific models and methods that have been applied in MPS dose reconstructions. The following topics are addressed:

- Methods of developing exposure scenarios and potential errors arising from different components of an exposure scenario
- Improved dose estimates through development of site-specific biokinetic models
- Reliability of bioassay data
- Reliability of air monitoring data

This article does not address the following topics concerning retrospective dose assessments for internal emitters that have been addressed frequently in the literature: methods of back-calculation of radionuclide intakes based on model fits to bioassay data, uncertainties in predictions of biokinetic models used in radiation protection or dose reconstruction, and uncertainties in dose estimates derived from bioassay data. The reader is referred to the following articles and reports for discussions and bibliographies on those topics (Leggett et al. 1998, Harrison et al. 2001, 2002; Leggett 2001; 2007; Boice et al. 2006; Bess et al. 2007; Pawel et al. 2007; NCRP 2009, 2018; Poudel et al. 2018).

Most of the illustrations in this article come from dose reconstructions performed for four MPS sites referred to as Rocketdyne (Boice et al. 2006), Mound (Boice et al. 2014), Mallinckrodt (Ellis et al. 2018), and Los Alamos National Laboratory (LANL) (Wiggs et al. 1994). Some illustrations from related sites studied by the authors or addressed in the literature are also provided.

Methods

This article consists largely of illustrations of the types, quantity, and quality of data available to derive retrospective dose estimates for internal emitters and some of the methods and models used to derive those estimates. The illustrations are taken mainly from monitoring data or other information collected for selected MPS sites. Four MPS sites are featured:

- Rocketdyne nuclear facility, near Los Angeles, California,
- Mound nuclear facility, Miamisburg, Ohio,
- Mallinckrodt Chemical Works, St. Louis, Missouri,
- LANL, Los Alamos, New Mexico.

The dose reconstructions for Rocketdyne, Mound, and Mallinckrodt have been completed and are summarized in the open literature (Boice et al. 2006, 2014, 2018; Ellis et al. 2018) and in a report of the National Council on Radiation Protection and Measurements (NCRP) (2018). Dose reconstructions for LANL workers are in progress at the time of preparation of this article; these will update and expand results of an earlier study (Wiggs et al. 1994). The general methods of dose reconstruction and evaluation of uncertainties in dose estimates in the MPS are described in detail in the NCRP report (NCRP 2018; Dauer et al. 2019).

Rocketdyne was involved in a wide range of radiological projects from 1948 to 1999, including uranium fuel fabrication, sodium-cooled breeder reactor technology, spent fuel evaluation, radiography, radiochemistry, plutonium (Pu) fuel fabrication, and storage of nuclear material. Most nuclear operations were ended by the early 1980s, the main exceptions being decladding of irradiated reactor fuel, decontamination and decommissioning of the nuclear facilities, storage of radioactive material, and applied physics experiments. Over 2200 workers were monitored for intake of radionuclides. Monitoring data are available for many different radionuclides including ^{234}U, ^{235}U, ^{238}U, ^{232}Th, ^{239}Pu, ^{241}Am, ^{210}Po, ^{137}Cs, and ^{90}Sr. The most important internal emitters at Rocketdyne were ^{234}U, ^{235}U, and ^{238}U (Leggett et al. 2005; Boice et al. 2006).

The worker cohort in the Mound study includes workers involved in a ^{210}Po project conducted at a Laboratory in Dayton, Ohio, from 1943 to 1948, when the project was transferred to the Mound site. The Dayton facility separated ^{210}Po from natural materials for use in polonium-beryllium neutron generators. This project was continued at Mound until 1959. Several other projects involving various radionuclides were conducted at Mound between 1949 and 1995, including production of ^{210}Po heat sources for radioisotope thermoelectric generators from 1953 to 1969 and ^{238}Pu heat sources from 1959 to 1995. Over 2300 workers had biossays

for polonium and about 1500 had bioassays for Pu (Boice et al. 2014).

The Mallinckrodt Chemical Works site was used from 1942 to 1957 for production of forms of uranium needed for the Manhattan Project. Purified uranium was produced by processing pitchblende from the Belgian Congo. Mallinckrodt operations included extraction of ^{226}Ra, because the company that provided the uranium ore retained ownership of its radium content. Other processes with high potential for intake of ^{238}U, ^{235}U, and their chain members included recovery of uranium from scrap uranium metal, machining of natural uranium metal rods to make reactor fuel slugs, reverting UF_4 to UO_2 or U_3O_8, production of UO_2F_2, and extraction and concentration of ^{230}Th from pitchblende raffinate. The most important internal emitters at Mallinckrodt appear to be the natural uranium isotopes and ^{226}Ra. Over 1900 workers had bioassays for uranium and about 500 had bioassays for ^{226}Ra (Ellis et al. 2018).

LANL has handled most natural or man-made radionuclides at one time or another, but mostly in small quantities. The most important source of dose to LANL workers from internal emitters is Pu, which has been handled in relatively large quantities at LANL since 1943. Early roles of LANL included reduction of Pu to metallic form, determination of its physical and metallurgical properties, and development of the technology required to build a nuclear bomb using Pu. Later programs have included peacetime applications of Pu such as development or testing of ^{238}Pu heat sources for space electric power generation or powering artificial organs. Over 1700 workers had positive bioassays for ^{239}Pu and nearly 600 had positive bioassays for ^{238}Pu.

For the most part, the biokinetic models applied in the dose reconstructions conducted to this point in the MPS are the models applied in ICRP Publication 68 (1994a). For the LANL study and for some special cases at Rocketdyne and Mound described in a later section of this article, the models of Publication 68 have been replaced with updated or site-specific biokinetic models. For most inhalation exposures at Rocketdyne, Mound, and Mallinckrodt, the applied respiratory model was the Human respiratory tract model (HRTM) introduced in ICRP Publication 66 (1994b) and applied in Publication 68. The ICRP originally provided three sets of parameter values for the HRTM describing different levels of solubility ('absorption types') of inhaled particles in the respiratory tract: Type F, representing fast dissolution and a high rate of absorption to blood; Type M, representing a moderate rate of dissolution and a moderate rate of absorption to blood; and Type S, representing slow dissolution and slow absorption to blood. These absorption types are referred to frequently in this article and, unless otherwise indicated, refer to their original definitions.

An updated version of the HRTM described in ICRP Publication 130 (2015) was applied to ^{239}Pu and ^{238}Pu inhalation cases in the LANL study. As discussed later, worker-specific parameter values were applied to Pu intakes in some cases.

Results

Importance of the exposure scenario

Definition

An exposure scenario used for a dose assessment of a known or suspected intake of a radionuclide is essentially a model describing the details of the exposure required to develop a meaningful dose estimate for a worker or a group of workers thought to have similar exposures. An exposure scenario for internally deposited activity includes the mode of intake (e.g. inhalation, wound, ingestion, or absorption through intact skin); the internally deposited radionuclide(s); the date(s) and pattern of intake (e.g. acute intake at a specific time, or chronic intake over a specified period); and the physical and chemical form(s) of the internally deposited radionuclide(s). Identification of the actual exposure conditions where feasible, or assignment of the most likely exposure conditions where information is incomplete, is a critical step toward derivation of meaningful retrospective dose estimates for an internal emitter.

Mode of intake

Bioassay records may provide clear evidence that a worker had an acute or extended intake of a radionuclide, or health physics records other than bioassay data may indicate that a worker was present in areas of high exposure to a radionuclide. The primary mode of the known or assumed intake often cannot be determined with much confidence from health physics records or available bioassay data. In the absence of specific information, it is generally assumed that intake was by inhalation because experience indicates that this is the most common mode of exposure to radionuclides in occupational settings. The second most common mode of intake at MPS sites studied thus far appears to be via wounds from contaminated objects. In a compilation of incidents at LANL with potential intake of ^{238}Pu or ^{239}Pu, roughly one-fifth of the cases involved wounds. Available incident reports for the Mound site suggest that some of the highest intakes of ^{210}Po may have been through wounds or absorption through intact skin. As illustrated in Figure 1, it is generally not possible to distinguish between an inhalation intake and a wound intake based on patterns of excretion of a radionuclide. The default assumption that internally deposited radionuclides entered the body in inhaled air can result in a large overestimate of the lung dose if this was not the actual intake mode or primary intake mode.

For dose estimates based on reasonably detailed urinary excretion data, the intake route tends to be a much less important source of uncertainty for estimated doses to systemic tissues than for estimated doses to tissues along the route of entry into the body, particularly the respiratory tract. This is because the rate of urinary excretion of a radionuclide does not definitively reveal the mode of intake or, for presumably inhaled activity, the distribution and retention time in the respiratory tract. Regardless of the route of intake, urinary activity is expected to reflect the level of activity that reached the systemic circulation and the characteristic

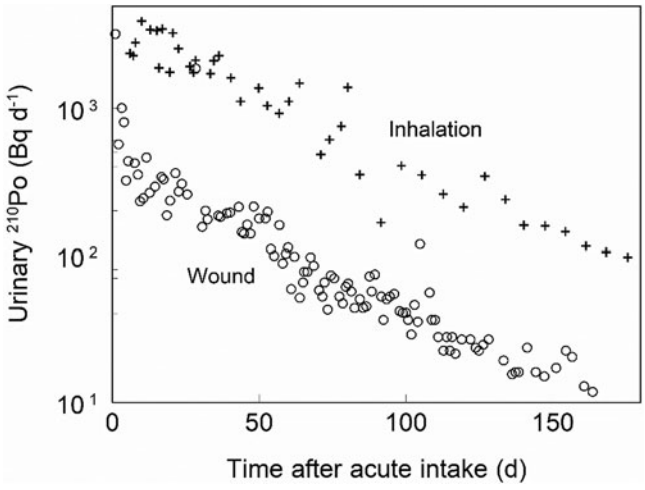

Figure 1. Patterns of time-dependent urinary excretion of ^{210}Po by two acutely exposed Mound workers, one via inhalation and the other via a contaminated wound.

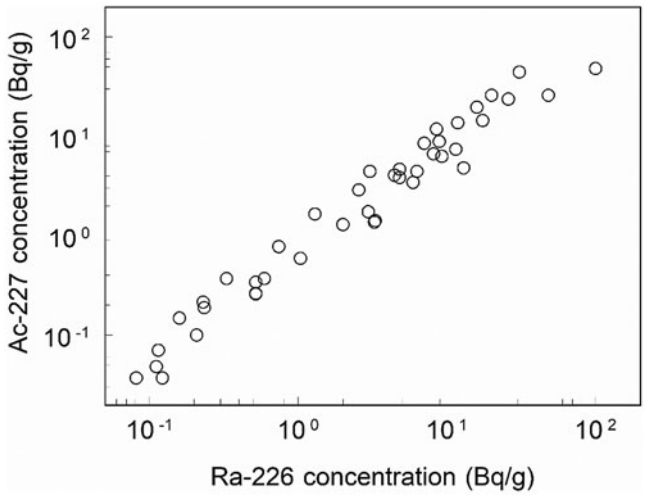

Figure 2. Relation of ^{226}Ra and ^{227}Ac concentrations in radioactive waste material originating at the Mallinckrodt site.

systemic kinetics of the radionuclide, with subject-specific variations due to biological variability.

Identification of internal emitters

As a practical matter, the internal emitters addressed in exposure scenarios and dose reconstructions for MPS sites usually have been only those that site health physicists considered sufficiently important to include in their internal dosimetry monitoring programs. As illustrated below, however, assessments of the uncertainty in estimated total doses for MPS sites should address radionuclides that were not monitored but perhaps represent important missed doses for some workers.

Much of the work performed at the Mallinckrodt site involved extraction of U from pitchblende ore and concentration of the extract (Ellis et al. 2018). The operations also included extraction of ^{226}Ra, which was required because the company that provided the ore retained ownership of its radium content. Bioassay programs at Mallinckrodt focused on intake of natural U isotopes, ^{226}Ra, and ^{222}Rn. Urinalyses for thorium, presumably for monitoring ^{230}Th intake, were also performed for a brief period. The air monitoring program focused on measurement of U isotopes, total alpha activity, and ^{222}Rn. The MPS dose reconstruction for Malinckrodt was limited to intake of U isotopes, ^{226}Ra, and ^{222}Rn and its short-lived progeny. There is also suggestive evidence of exposure of Mallinckrodt workers to the alpha emitter ^{227}Ac, which was not addressed in the dose reconstruction due to lack of monitoring data. Records indicate that ^{227}Ac may have been precipitated in high concentration in a process used to extract ^{226}Ra. Radiological surveys of a site that received residues from radionuclide separation processes at Mallinckrodt revealed similarly high concentrations of ^{226}Ra and ^{227}Ac in soil (Figure 2). A post-operation survey at Mallinckrodt also revealed hot spots in soil with high concentrations of both ^{226}Ra and ^{227}Ac. Such findings provide suggestive evidence that Mallinckrodt workers may have sometimes inhaled similar levels of ^{227}Ac and ^{226}Ra, but not sufficient evidence to warrant the assumption of intake of ^{227}Ac in a Mallinckrodt dose reconstruction performed for epidemiological purposes. Nevertheless, potentially missed doses from exposure to ^{227}Ac, which can yield far higher dose per inhaled Bq than ^{226}Ra, should be considered when addressing uncertainties in retrospective dose estimates for Mallinckrodt workers.

Dose reconstruction based on bioassay data often is complicated by a lack of specificity in the measurement technique. For example, bioassay records for Rocketdyne often describe measured activity in urine as 'mixed fission products' or 'total alpha.' In such cases, there may be no compelling reason to assume any specific mixture of fission products or alpha emitters, so that dose estimates require subjective choices of radionuclide mixtures or dosimetrically dominant radionuclides. In dose reconstructions for the Rocketdyne site, mixed fission products were often assumed to consist entirely of ^{90}Sr. The rationale was that exposures to fission products at Rocketdyne generally occurred when workers were handling equipment contaminated with aged fission products; the longest-lived fission products expected to be present were ^{90}Sr and ^{137}Cs; and in cases of acute exposure the urinary excretion curves often were reasonably consistent with a ^{90}Sr intake (Figure 3) but not ^{137}Cs intake. This approach may have substantially underestimated doses to soft tissues, or ^{90}Sr may have indeed been the dosimetrically dominant radionuclide. In any case, lack of identification of specific internal emitters resulted in non-trivial uncertainties in dose estimates for urinary activity identified in such broad terms as mixed fission products or total alpha.

Time pattern of intake

In many cases of accidental intake of radionuclides, the time of intake was recorded in an incident report and appeared to have been determined closely enough to introduce little or no uncertainty into the dose reconstruction. If an intake was not recognized at the time of occurrence but was revealed later by a routine bioassay measurement, the assumed time pattern of intake sometimes represented a significant or even dominant source of uncertainty in the dose

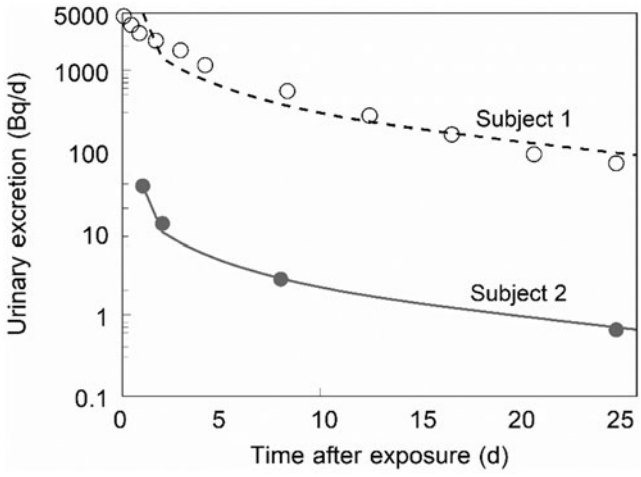

Figure 3. Urinary activity for two Rocketdyne workers, acutely exposed to mixed fission products at different times and locations. The indicated curve fits are based on intake of a relatively soluble form (Type F) of ^{90}Sr.

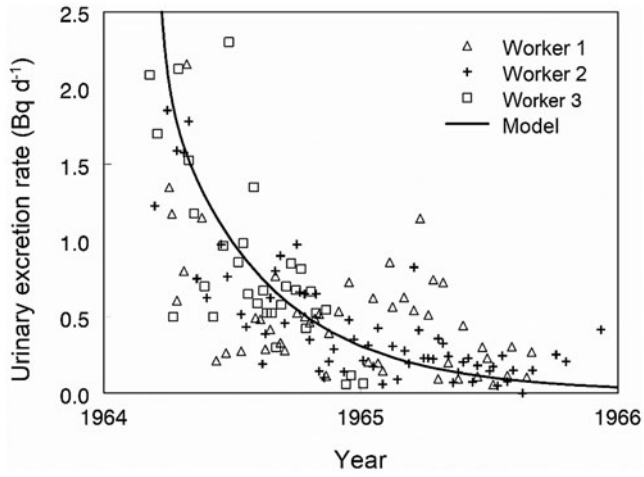

Figure 4. Urinary U data for three Rocketdyne workers acutely exposed to airborne U in the same incident. The model curve is based on acute inhalation of 10,000 Bq U, Type M, 5 μm AMAD.

estimate. For example, bioassay records for a Rocketdyne worker indicated intake of ^{239}Pu sometime within a 2.5-month period. Calculations based on different times of intake and a default form of inhaled Pu (Type M, particle size 5 μm AMAD) yielded estimates of ^{239}Pu intake ranging from 1 to 1200 Bq. Taking all sources of uncertainty into account including the form of inhaled material and errors in measurement of urinary ^{239}Pu, the uncertainty band on intake ranged from zero to several thousand Bq, with the time of intake being the dominant uncertainty in the dose estimate.

When bioassay data have indicated consistently low but nonzero intake of a radionuclide over an extended period, the standard assumption has been that the worker was chronically exposed at a constant rate. A time-dependent rate of intake has been applied to MPS sites when there is a sound basis for such a scenario, such as the existence of air monitoring data for uranium in the work area of interest.

LANL maintains a machine-readable database of incidents involving potential intake of ^{238}Pu or ^{239}Pu. For each incident and each worker involved in that incident, the database contains the date, worker identifier, and an abbreviation representing the nature of the exposure or reason for suspecting an intake (for example, w = wound, e = wound with excision, h = high nose count, r = high room count, u = unspecified accident). This database is a valuable aid in matching an increase in urinary ^{238}Pu or ^{239}Pu in a LANL worker with a likely time of intake. An incident database is particularly useful for dose reconstruction for Pu workers because intake of Pu appears to be associated more with incidents than with chronic exposure.

Form of an inhaled radionuclide

For inhalation exposure cases addressed in the MPS, the form of the inhaled material often can be reasonably well identified or inferred from health physics reports, information on the type of work performed, or other information. For example, intake of U at Rocketdyne was frequently associated with spontaneous uranium fires. For such cases, the intakes were generally assumed, and sometimes specified in reports, to be U oxide. For a sizable number of Rocketdyne workers, chronic exposure to relatively high levels of U occurred in a work area known as the powder room, where U aluminide (UAl_x) was produced. The urinary excretion curves, fecal to urinary excretion ratios, and external lungs measurements for workers exposed to U in the powder room generally followed a characteristic pattern for UAl_x (Leggett et al. 2005). Similarly, elevated intakes of ^{238}Pu at Mound or LANL often occurred during production or testing of ^{238}Pu heat sources, and the excretion curves for workers exposed to ^{238}Pu in those operations often followed a pattern characteristic of a form of ^{238}Pu used in those heat sources. In the above situations, information on the formation of the material together with bioassay data provided sufficient support for assigning a specific form of the radionuclide to the exposure scenario.

For most intakes known or suspected to be via inhalation, however, the choice of a respiratory model has been based on less compelling information. For the Rocketdyne site, for example, the ICRP's HRTM for Type M material was used as the default respiratory model for inhaled U based on patterns of urinary excretion of U by workers with known acute intakes, which in most cases were broadly consistent with inhalation of Type M material as illustrated in Figures 4–6. Similarly, bioassay data for Rocketdyne workers identified as exposure to ^{90}Sr, and sometimes as mixed fission products as indicated earlier, often were suggestive of inhalation of relatively soluble material (Figure 7), in which case Type F was used as the default absorption type for such exposures. Excretion data for inhaled ^{210}Po at the Mound site were not consistent with any of the ICRP's standard absorption types. A default absorption type for ^{210}Po inhalation at Mound was developed from site-specific data together with published data on the behavior of inhaled ^{210}Po in human subjects and laboratory animals.

A later section discusses the site-specific respiratory models applied to cases of inhalation of ^{210}Po at Mound, UAl_x at Rockedyne, and ^{238}Pu at LANL and Mound.

Application of data for coworkers (surrogate data)

In dose reconstructions for the sites addressed here, the situation sometimes arose that follow-up urinary excretion

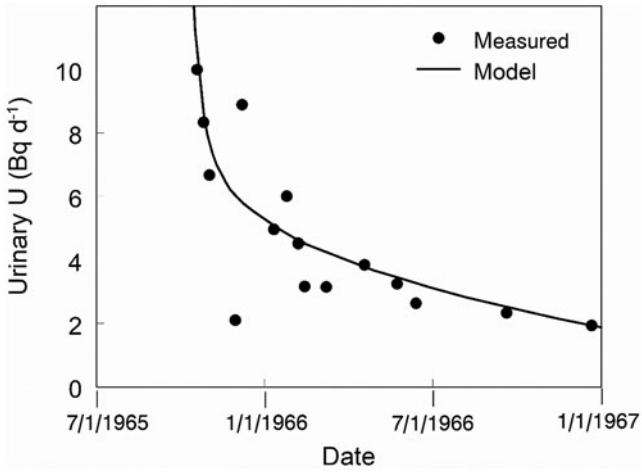

Figure 5. Observed and modeled urinary excretion rate for a worker acutely exposed to of U oxide in late 1965 and chronically exposed to lower quantities of U in 1966. The model predictions are based on inhalation of Type M material.

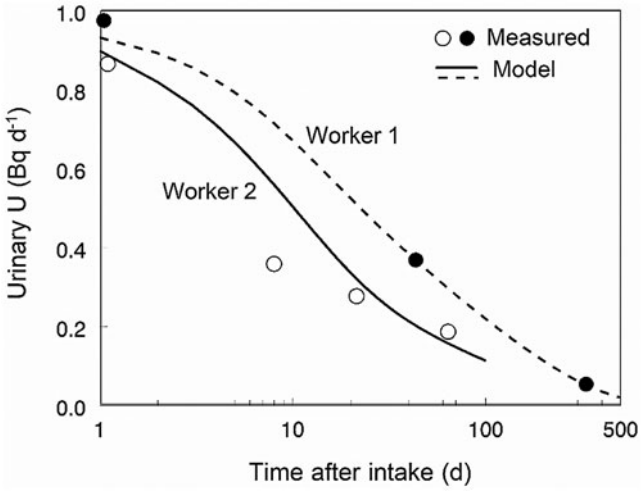

Figure 6. Observed and modeled urinary U for two workers acutely exposed to U at different times. Model predictions are based on inhalation of Type M material.

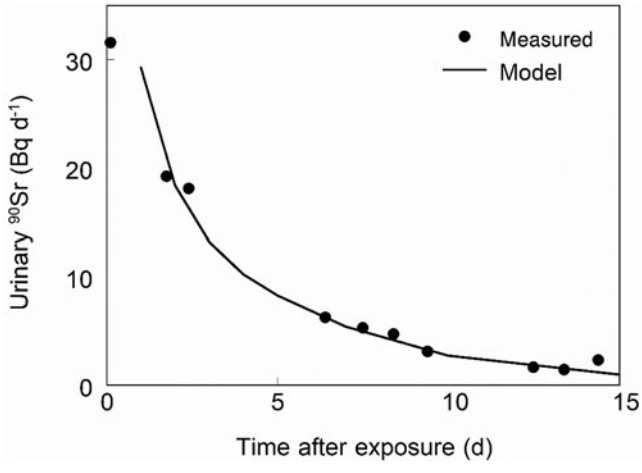

Figure 7. Observed and modeled urinary excretion rate for a worker presumed to have an acute intake of ^{90}Sr. The model predictions are based on inhalation of Type F material.

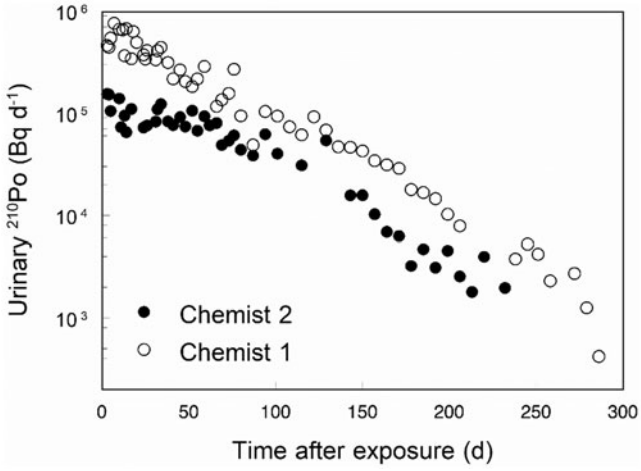

Figure 8. Urinary ^{210}Po data for two Mound chemists exposed to airborne ^{210}Po in the same incident.

measurements were available for some but not all workers listed in a report as being involved in an incident. In such cases, the same exposure scenario, but not necessarily the same level of intake, was assigned to each of the listed workers. In some cases, the radionuclide intake during the incident could be scaled from measurements of activity in nasal smears taken soon after the incident. If no data for nasal smears were available, a judgment had to be made as to whether to assign a zero intake to workers with no follow-up bioassay, or some positive intake extrapolated from data for workers with follow-up data. For example, if the incident occurred in a small, enclosed area, it seemed reasonable to assign an intake based on coworker data. If the incident occurred in a large open area, it seemed much less likely that all workers identified in the incident report had similar intake.

In any case, there is always considerable uncertainty in assigning intake data for a worker to an unmonitored coworker, as indicated by MPS case studies of follow-up monitoring of multiple workers involved in the same incident. For example, a Rocketdyne report lists 14 workers with potential intake of U resulting from a fire involving U carbide. As illustrated in Figure 4, three of the workers showed similar patterns and levels of urinary U over the following 1.5 years. The urinary excretion rate in a fourth worker was about a factor of 2 lower. The other 10 workers apparently had little intake of U.

In another case study, cumulative urinary ^{210}Po differed by a factor of 3 for two chemists at the Mound site who were acutely exposed in the same incident to high levels of airborne ^{210}Po (Figure 8).

In MPS dose reconstructions, common exposure scenarios sometimes have been applied to groups of workers based on simultaneous changes in their bioassay data. Common exposure scenarios have been applied to as few as two workers, such as the two Mound workers whose urinary ^{210}Po data are shown in Figure 9, and to relatively large groups of workers as illustrated by the following example. Bioassay records for the Rocketdyne site revealed an unusually large cluster of positive urinary U measurements for the first few weeks of 1963, with over 50

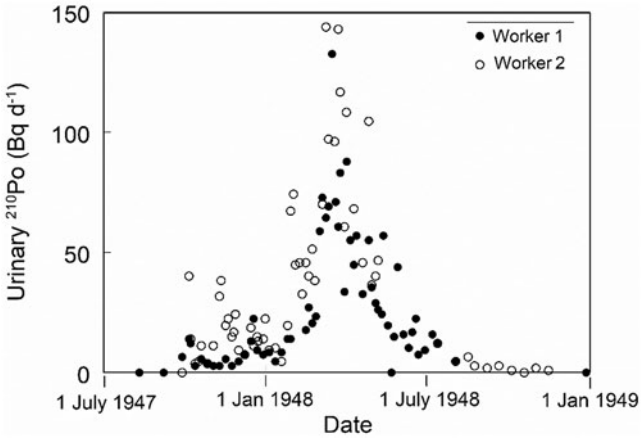

Figure 9. Similarity in urinary ^{210}Po patterns over a two-year period for two Mound workers.

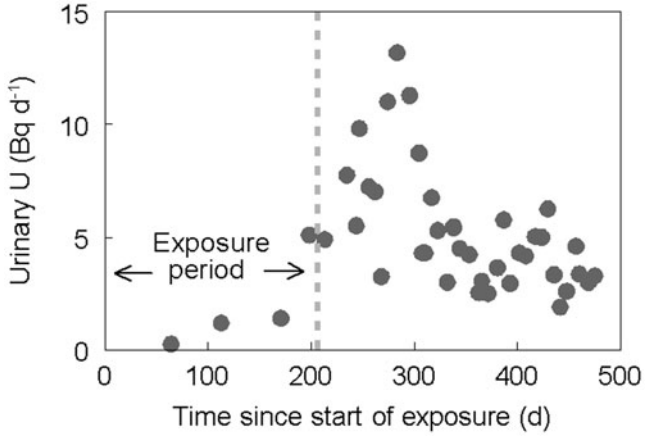

Figure 10. Urinary U data for a Rocketdyne worker exposed to airborne U aluminide.

workers showing elevated urinary U during that period. Similar patterns of decline in urinary U over the following months suggested an unrecognized short-term elevation of airborne U in a work area in early January 1963. In their dose reconstructions, these 50+ workers were all assumed to have been exposed acutely to a moderately soluble form of airborne U (Type M) on 2 January 1963. The level of intake of U for each worker was derived from the level of that worker's urinary excretion data.

As an aid in assigning exposure scenarios to groups of workers with little direct subject-specific information on exposure conditions, a history of known or suspected internal exposures to the Rocketdyne/AI workers was developed in the form of a time line. This is analogous to LANL's machine-readable database of incidents involving potential intake of ^{238}Pu or ^{239}Pu (described earlier) but contains verbal descriptions of exposures and includes some prolonged elevated intakes in addition to intakes resulting from incidents. This exposure time line was based on incident reports and bioassay data extracted from exposure histories of individual workers. Additionally, interviews of former radiation workers were conducted to learn directly from Rocketdyne workers about incidents, exposure circumstances, and work conditions. Incidents or prolonged elevated intakes involving a few hundred workers were identified. The largest single incident involved 35 workers exposed to a low level of airborne mixed fission products.

Illustrations of site-specific biokinetic models used in the MPS

Uranium aluminide intake at Rocketdyne

With a few exceptions, the highest estimated doses to Rocketdyne workers were for those involved in production of UAl_x, a material used in fuels for research and test reactors. Exposure to UAl_x occurred mainly in an area called the powder room, where UAl_x powder was formed and pressed into the cores of fuel plates. The UAl_x fuel fabrication program began in early 1966. Initially, monitoring consisted of widely spaced urinary U measurements and air sampling at fixed stations. Unexpected increases in urinary U were observed in some UAl_x workers in 1967. Subsequently, urinary excretion measurements were increased, the monitoring program was expanded to include fecal sampling and in vivo lung counting of ^{235}U, personal air monitoring was initiated, and remedial actions were taken to decrease airborne U.

The unexpected increase over time in the rate of urinary excretion of U presumably reflected an unusual pattern of behavior of UAl_x in the lungs. Initially, UAl_x deposited in the lungs dissolved at an extremely slow rate, but over time the inhaled particles apparently began to dissolve more rapidly, leading to a rapidly increasing rate of urinary excretion of U. The pattern of excretion relative to the time of exposure is illustrated in Figure 10, which shows data for a worker who was removed from exposure after a routine urine measurement revealed a large increase in urinary U since the previous measurement. His urinary U levels continued to increase for a few months after the end of exposure and then declined sharply. Because U is typically excreted at a high rate after absorption to blood and the rate of absorption from lungs to blood depends on the rate of dissolution in the lungs, the data indicate a substantial increase over time in the dissolution rate in the lungs.

Because none of the ICRP's default absorption types were applicable to UAl_x, special parameter values of the ICRP's HRTM were developed to describe the respiratory behavior of this unusual form of U (Leggett et al. 2005). The parameter values were chosen to reproduce as closely as feasible the urinary and fecal excretion patterns of U indicated by extensive bioassay data for the relatively large set of workers exposed to UAl_x and the clearance rate from the lungs as indicated by in vivo lung counts for UAl_x workers. A particle size of 1 μm AMAD rather than the default size of 5 μm AMAD was assumed for inhaled UAl_x based on information obtained from interviews of workers who had been involved in its production.

Figure 11 illustrates the fit of the UAl_x model predictions to urinary and fecal excretion data for a worker who was exposed to UAl_x for a few months. The exposure scenario is based on the relative change with time in the concentration of U in air in the powder room. That is, the rate of intake of UAl_x was assumed to be proportional to the time-dependent concentration of UAl_x in air throughout the exposure period. The absolute air concentrations of UAl_x were not used in the dose reconstruction for powder room workers.

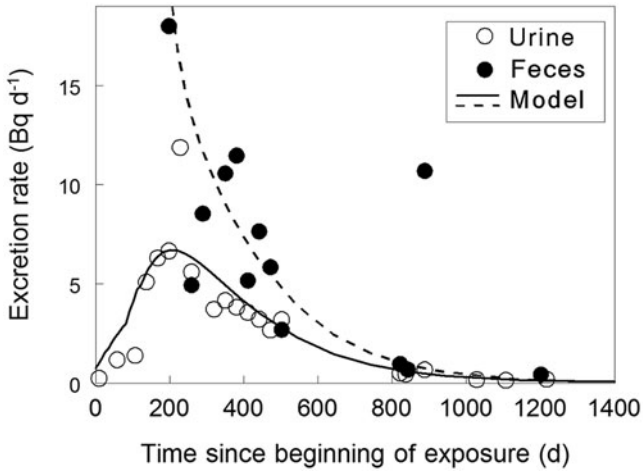

Figure 11. Model fit to urinary and fecal excretion data for a worker exposed to UAl_x.

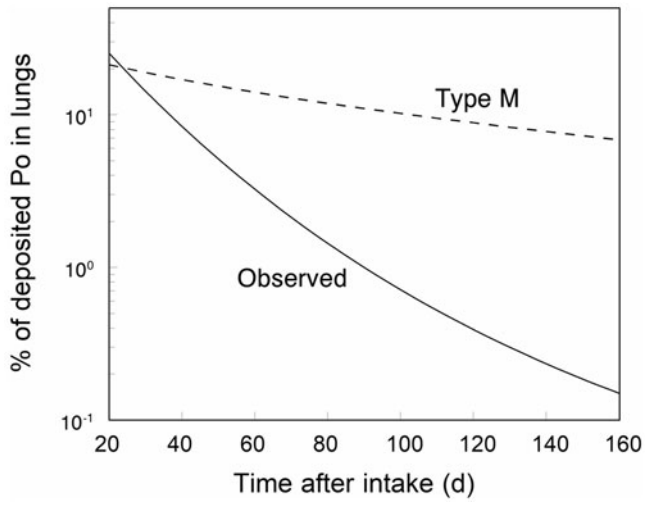

Figure 12. Lung retention of an inhaled ^{210}Po aerosol in dogs (curve labeled "observed" based on data of Smith et al. 1961), compared with predictions based on absorption Type M.

Polonium-210 intake at Mound

At the time of the MPS dose reconstruction for Mound, the ICRP's systemic model for internally deposited polonium was a model adopted in ICRP Publication 67 (1993), and the ICRP's default absorption type for inhaled polonium was Type M. Different models were applied in MPS dose reconstructions for ^{210}Po intakes at Mound. A systemic model of Leggett and Eckerman (2001) was applied. The ICRP's parameter values for Type M material were modified for consistency with patterns of excretion of ^{210}Po for Mound workers as well as consistency with published data on the behavior of inhaled ^{210}Po in the respiratory tract. Data for laboratory animals (Figure 12) and accidentally exposed Mound workers (Figure 13) suggested that Type M parameter values substantially overestimated the retention time of Po in the lungs for the form of Po likely to be encountered at Mound. Also, data for a worker exposed to ^{210}Po oxide, presumably one of the less soluble forms of polonium, at another site (Scott and West 1975) indicate much faster removal from the lungs than depicted by Type M parameter values. For derivation of doses from inhaled ^{210}Po, the long-term biological half-time of ~140 d for polonium in the deep lungs specified in ICRP Publication 66 for Type M material was replaced with a half-time of 30 d. A particle size of 1 μm AMAD was assumed for inhaled polonium rather than the default size of 5 μm AMAD because airborne ^{210}Po seemed likely to exist as fine aerosols in view of the types of operations conducted at Mound.

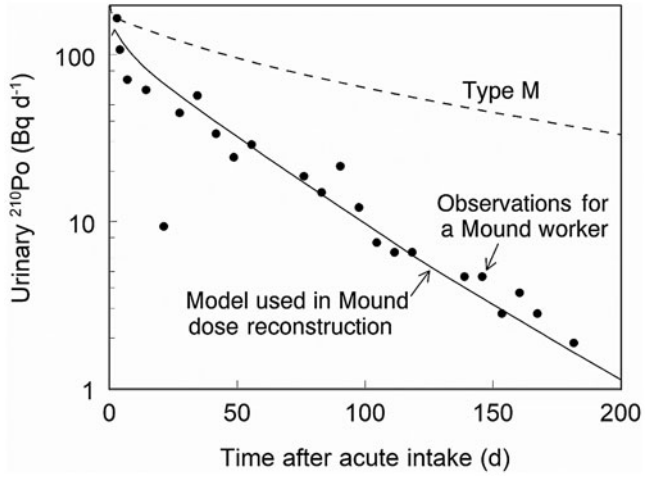

Figure 13. Model predictions of rate of decline of urinary ^{210}Po following inhalation, based on alternate respiratory models, compared with data (close circles) representative of the typical pattern of excretion of inhaled ^{210}Po in Mound workers.

Radium-226 intake at Mallinckrodt

A non-standard approach to modeling intake of ^{226}Ra at Mallinckrodt arose from the type of bioassay data used to monitor ^{226}Ra exposure, rather than from specific information on the chemical or physical form of inhaled ^{226}Ra.

Exposure to ^{226}Ra was monitored at Mallinckrodt through measurement of exhaled ^{222}Rn rather than its rate of loss in urine. Measurement of ^{222}Rn in breath was made at Mallinckrodt during the period 1948–1959. Figure 14 shows ^{222}Rn breath measurements for two Mallinckrodt workers.

The biokinetic and dosimetric models of ICRP Publication 68 (1994a) were used to estimate intake of ^{226}Ra for a worker based on breath ^{222}Rn data, assuming all ^{226}Ra intake was via inhalation of a moderately soluble form (Type M) of particle size 5 μm AMAD. It was assumed in the MPS dose reconstruction for Mallinckrodt workers that the integrated ^{222}Rn activity in breath over the entire monitoring period for a radium worker was proportional to the integrated ^{226}Ra in the body over that period, and the ^{226}Ra intake rate was constant over that period. The constant ^{226}Ra intake rate for an individual worker could then be derived using the biokinetic models of ICRP Publication 68, the worker's continuous breath ^{222}Rn curve, and a conversion factor that relates breath ^{222}Rn to a typical body content of ^{226}Ra over an extended period.

To convert from ^{222}Rn in breath to ^{226}Ra in the body it was assumed that 1 Bq ^{222}Rn per liter of air exhaled during a breath test corresponds to a body ^{226}Ra content of 72,000 Bq. This relation is derived from a formula proposed

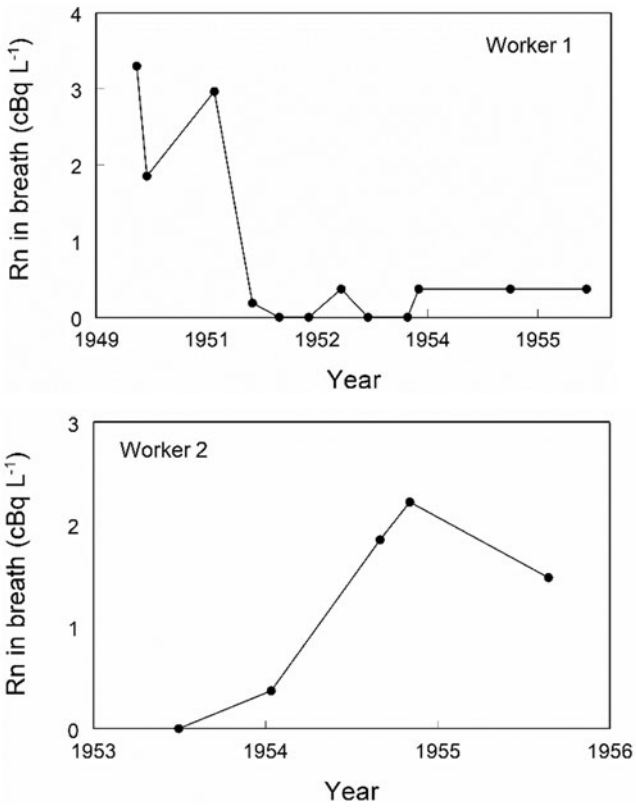

Figure 14. Measurements of ^{222}Rn in breath in two Mallinckrodt workers exposed to ^{226}Ra.

by Srivastava et al. (1986) and is reasonably consistent with predictions of the biokinetic models used in this study assuming extended exposure to airborne ^{226}Ra. Srivastava and coworkers concluded that the ^{226}Ra body burden could be estimated from the rate of exhalation of ^{222}Rn by the relationship

$$^{226}\text{Ra in the body (Bq)} = (\text{C BR})/(\lambda \text{F}) \quad (1)$$

where BR is the breathing rate in liters (h^{-1}), λ is the decay constant of ^{222}Rn (h^{-1}), C is the concentration of ^{222}Rn in the breath sample (Bq L^{-1}), and F is the fraction of ^{222}Rn produced in the body that is exhaled. Srivastava et al. estimated a typical release fraction F of 0.84 and a range of F of 0.7–1.0, depending on whether ^{226}Ra is mostly in bone (relatively low F) or in lungs and other soft tissues. The relationship used in the MPS (1 Bq ^{222}Rn/L = 72,000 Bq ^{226}Ra) yields a lower estimate of body ^{226}Ra than generally estimated by this method because a relatively high breathing rate, representing a worker during a work period, and a relatively low value of F (down to 0.63) generally have been used. The relationship between radon exhalation and radium body burden used in the MPS dose reconstruction considers that the workers were resting during the breath tests and had a lower breathing rate than the reference value for a worker.

There are several sources of uncertainty in the reconstructed doses for ^{226}Ra intakes by Mallinckrodt workers, including the cumulative ^{222}Rn exhalation, the relation of ^{222}Rn in breath to the content of ^{226}Ra in the body, the solubility of inhaled ^{226}Ra from the lungs, and the possibility of ^{226}Ra intake via other pathways including wounds and ingestion. Overall, the largest uncertainties may be associated with the estimate of cumulative ^{222}Rn in breath, in view of the high variability in the ^{222}Rn exhalation rate and the fact that a worker typically had only 1–4 breath tests per year.

Plutonium-238 intake at LANL

The production of ^{238}Pu heat sources was started at Mound in 1961. Determination of properties of the heat sources and some production support began at LANL in the late 1960s. Production of ^{238}Pu heat sources was largely transferred to LANL by the late 1970s. Published data and internal incident reports, together with bioassay data for the workers involved, indicate a sizable number of acute LANL to the high-fired ^{238}Pu dioxide used in ^{238}Pu heat sources. Generally, the observed pattern of urinary excretion of activity following inhalation of ^{238}Pu dioxide is broadly similar to the pattern described above for inhaled UAl_x. That is, there is initially little urinary excretion of activity after exposure to relatively high concentrations of airborne ^{238}Pu, but the inhaled material begins to dissolve more rapidly in the lungs over an extended period and subsequently is excreted at a substantially increased rate. Daily loss of ^{238}Pu in urine typically peaks at least a few months, and up to a few years in some cases, after acute intake and then declines gradually thereafter.

Dose estimates for inhaled ^{238}Pu dioxide at LANL are based on worker-specific variations of respiratory parameter values for ^{238}Pu dioxide that have been adopted by the ICRP for use in an upcoming report (Part 4 of an ICRP series on occupational intake of radionuclides, in press). A similar but not identical model with worker-specific parameter values was applied to ^{238}Pu dioxide intakes at Mound, prior to the completion of the ICRP's ^{238}Pu dioxide model. The worker-specific parameter values are set to reproduce the time and level of peak urinary excretion of ^{238}Pu following acute inhalation of ^{238}Pu dioxide. Figure 15 shows model fits to ^{238}Pu excretion data for two LANL workers and two Mound workers acutely exposed to airborne ^{238}Pu-dioxide.

Illustrations of problematic bioassay data

Two types of uncertainty associated with bioassay data

The components of uncertainty in a bioassay measurement may be divided into two main categories commonly referred to as Type A and Type B uncertainties (ICRP 2015). Type A uncertainties are those arising from counting statistics, and Type B components are those associated with all other sources of uncertainty. Type A uncertainties tend to decrease with increasing activity or counting time, while uncertainties associated with Type B may be largely independent of activity or counting time. Type B uncertainties associated with measurement of activity in a urine sample, for example, would include uncertainties in sample volume, calibration, recovery, background, contamination, and conversion of results for a spot sample to a 24-h excretion rate.

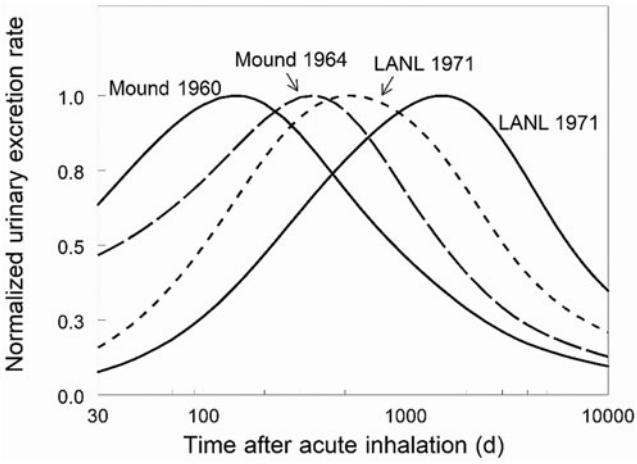

Figure 15. Differences in peak urinary excretion times for workers acutely exposed to ^{238}Pu dioxide. Each curve is a model fit to urinary ^{238}Pu data for an individual worker.

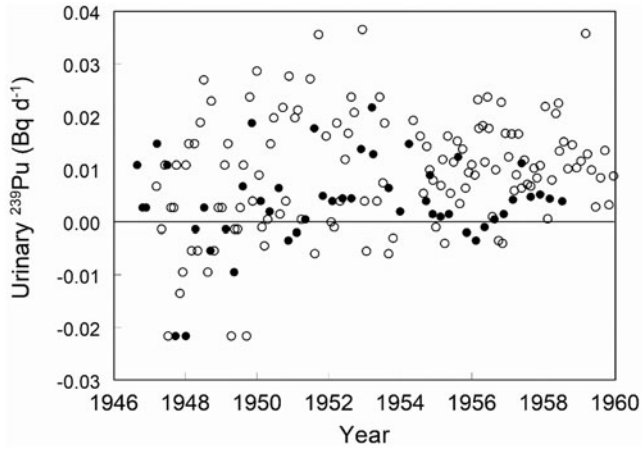

Figure 16. Urinary ^{239}Pu measurements for two LANL workers, illustrating a reduction over time in the scatter of measurements and the magnitude of negative results.

The illustrations in this section address some important uncertainties (largely Type B) that have been encountered in MPS dose reconstructions in interpretation of urinary excretion data, because this is the type of bioassay data most commonly available for MPS sites.

Uncertainties in urinary Pu data for LANL workers

Urinary Pu measurements frequently involve important Type A uncertainties due to the low rate of excretion of Pu following its uptake to blood. The following brief history of the urinary Pu measurement program at LANL during the 1940s and 1950s (Campbell et al. 1972; Hempelmann et al. 1973; Lawrence 1978; Miller et al. 2008) describes some important Type B uncertainties such as high background (blank) counts and contamination issues that, together with common counting issues for urinary Pu, appear to have resulted in unreliable urinary Pu measurements in the early years of the LANL bioassay program.

A program for measurement of urinary activity in Pu workers was instituted at LANL in 1944. Twenty-four-hour urine samples were collected, with all urine voided by a worker during the day collected in a single bottle in presumably clean areas after decontamination of the worker and a change of clothes. The bottle was taken home at night and returned the next morning to complete the 24-h collection. Occasional extremely high activities suggested contamination problems, perhaps ^{210}Po contamination in some cases as Pu and Po were handled in the same work area. New methods were started in 1945 to reduce contamination issues. Workers were given two paid days to be spent away from work and away from Los Alamos, after which the workers reported to a hospital where they provided urine samples in an environment that was kept as clean as feasible.

From 1944 to 1957, LANL used the cupferron method of urinary Pu measurement in which Pu with an iron carrier was extracted by cupferron, a precipitant for iron and copper. The original counting system had high background counts that made it difficult to achieve a meaningful determination of the concentration of Pu in urine because of the low rate of excretion of absorbed Pu. The counting system was replaced in 1945 with a system that reduced background counts by as much as a factor of 30, and later improvements further reduced background counts by another order of magnitude. Figure 16 shows urinary ^{239}Pu measurements for two LANL workers, both involved in multiple incidents in the 1940s with apparent intake of ^{239}Pu. The figure illustrates a decrease over time in data scatter and in the magnitude of negative results for early LANL worker, presumably reflecting improved measurements due to reduced background counts.

In 1948, the two-day vacation system was abandoned due to the expense. The only time off work for purposes of limiting contamination of urine samples was the 1 d during which the sample was collected in the hospital. Urine collection methods were further simplified in 1952 to allow relatively frequent sampling of a larger group of workers and again in 1958 to solicit more favorable employee cooperation. The method for determining the concentration of Pu in urine was updated in 1957, when the cupferron method was replaced by the nuclear-tract method of alpha counting (NTA method) in an effort to increase the sensitivity of the bioassay procedure. Further improvements were made frequently thereafter.

To summarize, it appears that urinary Pu measurements during the first year or so of the bioassay program at LANL involve large uncertainties due to high background counts and high potential for contamination of samples. Considerable improvements were made in 1945 regarding reduction of background counts and contamination, but the reliability of the improved method is difficult to quantify. The potential for contamination of urine samples may have increased again in the late 1940s and early 1950s due to efforts to monitor more workers more frequently and encourage workers to cooperate with the bioassay program. A considerable improvement in the Pu assay methods apparently was made in 1957, when a more sensitive measurement technique was introduced.

Dominant uncertainty in urinary ^{210}Po data for Mound workers

An important uncertainty in reconstruction of doses from intake of ^{210}Po by Mound workers was the level of recovery of

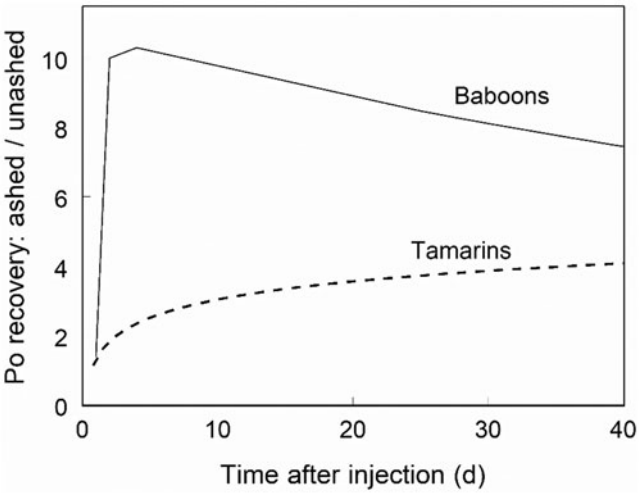

Figure 17. For different primates, the time-dependent increase in recovery of ^{210}Po from urine resulting from wet ashing of urine before measurement of ^{210}Po by the technique used at Mound.

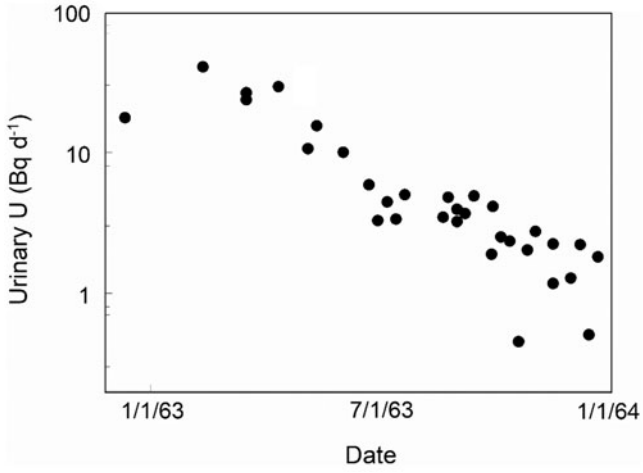

Figure 18. Urinary U levels in a worker at a U fuel production plant that had relied solely on air monitoring for the previous two years.

^{210}Po from urine (a Type B uncertainty). The technique used at Mound to measure the ^{210}Po concentration in urine samples involved spontaneous deposition of ^{210}Po onto a metal disc from which its decays were counted. An issue apparently not recognized until after completion of the ^{210}Po program at Mound was that Po excreted in urine may not be recovered to the same extent as tracer Po added to urine to determine the fraction recovered, unless there is wet ashing (acid digestion) of the urine prior to deposition on the disc. The Mound technique did not involve wet ashing of the urine. Results of human and animal studies of the percentage of excreted polonium recovered from unashed urine are highly variable (Black 1956; Sedlet and Robinson 1971; Bale et al. 1975; Fellman et al. 1989, 1994). The available data suggest that the increase in recovery of Po from urine due to wet ashing may vary with time after intake and with animal species. For example, with the measurement technique applied at Mound, wet ashing increased Po recovery from urine by a factor of 7–10 in baboons (excluding day 1), depending on time after intake, and a factor of about 2–4 in tamarins, depending on time after intake (Figure 17). In the MPS dose reconstruction for Mound, it was assumed that that recovery was 20%, a best estimate based on the collective data. This applied value might have overestimated actual recovery by a factor of 2 or underestimated recovery by a factor of 3 or more.

Limitations of air monitoring data

Air monitoring can provide a useful warning system for release of radionuclides into a work area, but measured air concentrations of radionuclides often are crude and unreliable predictors of intake. This conclusion is based on: (1) comparisons of intake derived from air monitoring data with intake based on bioassay data at MPS sites (e.g. in the Rocketdyne 'powder room' where UAl_x was produced) as well as other facilities (Britcher and Strong 1994; Britcher et al. 1998; Eckerman and Kerr 1999) and (2) the fact that different methods of air monitoring often yield considerably different results (Marshall and Stevens 1980; Britcher and Strong 1994).

Marshall and Stevens (1980) found that concentrations of airborne radionuclides based on personal air samplers (PAS) can be up to 50 times greater than values based on static air samplers (SAS). Britcher and Strong (1994) concluded from a review of monitoring data for a group of workers in the UK that intakes assessed from PAS data were about an order of magnitude greater than those implied by SAS data. In a review of the use of PAS as part of internal dosimetry monitoring programs at two facilities in the UK, Britcher and Strong (1994) concluded that the correlation of radionuclide intake between assessments using PAS and biological samples was poor. Both SAS and PAS data appear to be particularly unreliable for reconstructing acute intakes of radionuclides. Estimation of radionuclide intake based on air sampling is further complicated by inconsistent use of masks, which can reduce intake by as much as a factor of 50. For these reasons, air monitoring data generally have not been used in the MPS to derive intake of radionuclides except for radon measurement or as an indicator of relative intakes of a radionuclide over an extended period.

The following example describes a failure in an air monitoring system at a U.S. facility (not an MPS site) that produced U metals and compounds from natural and enriched uranium feed stocks for use as fuel in nuclear reactors. Routine urinary U measurements were started in 1957 to supplement an air monitoring program. This bioassay program was discontinued in early 1961, but reinstated in late 1962 at the direction of the U.S. Atomic Energy Commission. The first set of urinary U measurements after the restart of the bioassay program revealed far higher intakes of U than had been indicated by air sampling, in an area where enriched U was handled. The highest intakes were associated with milling of UO_2. Urinary U data for one of the highly exposed workers are shown in Figure 18. Based on the assumption that urinary excretion data represent inhaled U, the estimated lung dose was at least 0.1 Sv assuming intake of Type M material. This could be a substantial

underestimate if much of the intake occurred long before the restart of the bioassay program or if the inhaled material was less soluble than assumed.

Conclusions

1. Much of the dose reconstruction effort for internal emitters should be devoted to development of best feasible exposure scenarios, including the time pattern and mode(s) of intake, the internally deposited radionuclide(s), and the form(s) of the internally deposited radionuclide(s).
2. Coworker data should be used to assign exposure scenarios or dose estimates to workers with missing exposure data if, and only if, there is compelling evidence of similar coworker exposure.
3. Bioassay data for some radionuclides and periods of operation at MPS sites are of questionable reliability due to sizable uncertainties associated with contamination, recovery, or background issues.
4. Dose estimates derived solely from air monitoring data should be treated as highly uncertain values in the absence of information demonstrating that the data are reasonably predictive of intake.
5. For intakes known or assumed to be via inhalation, the uncertainty in lung dose typically is much greater than the uncertainty in dose to systemic tissues, when dose estimates are based on urinary excretion data.
6. The lung dose estimate often can be improved through development of site-specific respiratory absorption parameter values.
7. There is generally insufficient site-specific information to justify development of site-specific systemic models. However, it may be justified to replace outdated ICRP models with more recently developed models.

Notes on contributors

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Keith F. Eckerman retired in 2013 from Oak Ridge National Laboratory and is an emeritus member of the National Council on Radiation Protection (NCRP) and Committee 2 of the International Commission on Radiological Protection (ICRP). He has over 40 years' experience in radiation protection at Argonne National Laboratory, the Nuclear Regulatory Commission, and Oak Ridge National Laboratory. He served on ICRP Committee 2 for over 20 years and chaired their task group on dose calculations. He received the Health Physics Society Distinguished Scientific Achievements Award in 1995, the Loevinger-Berman Award of the Society of Nuclear Medicine in 2001, the Radiological Protection Gold Medal from Royal Swedish Academy of Sciences in 2012, and Lauriston S. Taylor award of the NCRP in 2015. He has a PhD in radiological physics from Northwestern University.

Dr. ***Michael Bellamy*** is a nuclear engineer and health physics researcher at the Oak Ridge National Laboratory in the United States. He has served for over 10 years at the Center for Radiation Protection Knowledge, specializing in Monte Carlo radiation transport research and internal dosimetry modeling. He is currently focused on internal dose reconstruction for the MPS. Dr. Bellamy's research supports several federal entities including the Centers for Disease Control and Prevention, the U.S. Department of Energy, the U.S. Nuclear Regulatory Commission, and the U.S. Environmental Protection Agency.

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